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(REV 10-95)

U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

ATTORNEY'S DOCKET NUMBER

**TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. §371**

MERCK 2192

U.S. APPLICATION NO. (if known, see 37 C.F.R. §1.5)

097/743758

INTERNATIONAL APPLICATION NO.

INTERNATIONAL FILING DATE

PCT/EP99/04792

8 JULY 1999

PRIORITY DATE CLAIMED

16 JULY 1998

TITLE OF INVENTION

COLORING USING PEARLESCENT PIGMENTS IN THE FOOD AND PHARMACEUTICAL SECTORS

APPLICANT(S) FOR DO/EO/US

SCHWEINFURTH, Ralf, et al.

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. §371.
 2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. §371.
 3. ☐ This express request to begin national examination procedures (35 U.S.C. §371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. §371(b) and PCT Articles 22 and 39(1).
 4. ☒ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
 5. ☒ A copy of the International Application as filed (35 U.S.C. §371(c)(2))
 - a. ☐ is transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☒ has been transmitted by the International Bureau.
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
 6. ☒ A translation of the International Application into English (35 U.S.C. §371(c)(2)).
 7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. §371(c)(3))
 - a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☐ have been transmitted by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☒ have not been made and will not be made.
 8. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. §371(c)(3)).
 9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. §371(c)(4)).
 10. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. §371(c)(5)).
- Items 11. to 16. below concern document(s) or information included:**
11. ☐ An Information Disclosure Statement under 37 C.F.R. §§1.97 and 1.98.
 12. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 C.F.R. §§3.28 and 3.31 is included.
 13. ☒ A **FIRST** preliminary amendment.
 - ☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
 14. ☐ A substitute specification.
 15. ☐ A change of power of attorney and/or address letter.
 16. ☐ Other items or information:

U.S. APPLICATION NO. (if not, see 37 C.F.R. § 1.53)		INTERNATIONAL APPLICATION NO.		ATTORNEY'S DOCKET NUMBER	
09/743758		PCT/EP99/04792		MERCK 2192	
17. <input checked="" type="checkbox"/> The following fees are submitted:				CALCULATIONS PTO USE ONLY	
BASIC NATIONAL FEE (37 CFR §1.492 (a) (1) - (5)): Search Report has been prepared by the EPO or JPO..... \$860.00 International preliminary examination fee paid to USPTO (37 CFR §1.482)..... \$690.00 No international preliminary examination fee paid to USPTO (37 CFR §1.482) but international search fee paid to USPTO (37 CFR §1.445(a)(2))..... \$710.00 Neither international preliminary examination fee (37 CFR §1.482) nor international search fee (37 CFR §1.445(a)(2)) paid to USPTO..... \$1000.00 International preliminary examination fee paid to USPTO (37 CFR §1.482) and all claims satisfied provisions of PCT Article 33(2)-(4)..... \$100.00					
ENTER APPROPRIATE BASIC FEE AMOUNT =				\$860.00	
Surcharge of \$130.00 for furnishing the oath or declaration later than months from the earliest claimed priority date (37 C.F.R. §1.492(e)). <input type="checkbox"/> 20 <input type="checkbox"/> 30					
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		
Total claims	11 - 20 =	0	x \$ 18.00	\$0.00	
Independent claims	3 - 3 =	0	x \$ 80.00	\$0.00	
MULTIPLE DEPENDENT CLAIM(S) (if applicable)			+ \$ 270.00		
TOTAL OF ABOVE CALCULATIONS =				\$860.00	
Reduction of 1/2 for filing by small entity, if applicable. A Verified Small Entity Statement must also be filed (Note 37 C.F.R. §§1.9, 1.27, 1.28).					
SUBTOTAL =				\$860.00	
Processing fee of \$130.00 for furnishing the English translation later than months from the earliest claimed priority date (37 C.F.R. §1.492(f)). <input type="checkbox"/> 20 <input type="checkbox"/> 30					
TOTAL NATIONAL FEE =				\$860.00	
Fee for recording the enclosed assignment (37 C.F.R. §1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 C.F.R. §§3.28, 3.31). \$40.00 per property.					
TOTAL FEES ENCLOSED =				\$860.00	
				Amount to be refunded:	
				charged:	
a. <input checked="" type="checkbox"/> A check in the amount of \$860.00 to cover the above fees is enclosed. b. <input type="checkbox"/> Please charge my Deposit Account No. 13-3402 in the amount of \$_____ to cover the above fees. A duplicate copy of this sheet is enclosed. c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 13-3402. A duplicate copy of this sheet is enclosed.					
NOTE: Where an appropriate time limit under 37 C.F.R. §§1.494 or 1.495 has not been met, a petition to revive (37 C.F.R. §1.137(a) or (b)) must be filed and granted to restore the application to pending status.					
SEND ALL CORRESPONDENCE TO:					
MILLEN, WHITE, ZELANO & BRANIGAN, P.C. Arlington Courthouse Plaza I 2200 Clarendon Boulevard, Suite 1400 Arlington, Virginia 22201 (703) 243-6333					
Filed: 16 JANUARY 2001				SIGNATURE	
AJZ:jmm				Anthony J. Zelano	
				NAME	
				27,969	
				REGISTRATION NUMBER	



23599

PATENT TRADEMARK OFFICE

IN THE UNITED STATES DESIGNATED/ELECTED OFFICE

International Application No. : PCT/EP99/04792
International Filing Date : 8 JULY 1999
Priority Date(s) Claimed : 16 JULY 1998
Applicant(s) (DO/EO/US) : SCHWEINFURTH, Ralf, et al.
Title: COLORING USING PEARLESCENT PIGMENTS IN THE FOOD AND PHARMACEUTICAL SECTORS

PRELIMINARY AMENDMENT

Commissioner for Patents
Washington, D.C. 20231

SIR:

Prior to calculating the national fee, and prior to examination in the National Phase of the above-identified International application, please amend as follows:

IN THE CLAIMS:

- Claim 3, line 2, delete "or 2";
- Claim 4, line 2, delete "one of Claims 1 to 3" and insert --Claim 1--;
- Claim 5, line 2, delete "one of Claims 1 to 4" and insert --Claim 1--;
- Claim 6, line 2, delete "one of Claims 1 to 5" and insert --Claim 1--;
- Claim 8, line 2, delete "or 7";
- Claim 9, line 2, delete "one of Claims 1 to 8" and insert --Claim 1--.

REMARKS

The purpose of this Preliminary Amendment is to eliminate multiple dependent claims in order to avoid the additional fee. Applicants reserve the right to reintroduce claims to canceled combined subject matter.

Respectfully submitted,



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Colouring using pearlescent pigments in the food and
pharmaceutical sectors

5 The present invention relates to the use of titanium dioxide pigments and/or iron oxide pigments based on platy substrates for colouring food products and pharmaceutical products.

10 In addition to their functional applications, pearlescent pigments and interference pigments are also increasingly being used for the visual enhancement of products, for example in cosmetics, since fine colours and effects give rise to pleasing subjective impressions on the part of the viewer and consumer.
15 Since, in the production of pearlescent pigments, for example for decorative cosmetics, the strictest requirements are made as to the purity and quality of the pigments, pearlescent pigments should also be usable in the food sector for improving the colouring
20 effect or for colouring.

The object of the present invention is therefore to expand the palette of the previously known colours in the colouring of food products and pharmaceutical
25 products using pearlescent pigments or interference pigments, as the result of which the products experience an additional sensorially perceptible enhancement. The visual enhancement is valuable in particular for pharmaceutical products, since a clearer
30 differentiation is made possible between differently coloured tablets, dragees, etc.

Surprisingly, it has now been found that for the visual enhancement of food products and pharmaceutical
35 products, platy substrates coated with titanium dioxide and/or iron oxide are outstandingly suitable. Iron oxide pigment is taken to mean platy substrates coated not only with Fe_2O_3 but also with Fe_3O_4 . In particular, the combination of TiO_2 pigments and/or Fe_3O_4 pigments

with natural or nature-identical dyes, colour pigments or colorant fruit extracts and plant extracts give the food product an interesting new colour. By means of the colouring of food products, simultaneously desires for
5 novel variants and nuances in colours as the result of novel trends in fashion can be taken into account.

The invention thus relates to the use of titanium dioxide pigments and/or iron oxide pigments based on
10 platy substrates for colouring food products and pharmaceutical products.

The coloured foods and pharmaceutical products are distinguished by a novel colouring effect which is
15 based on light refraction by the pearlescent pigments and causes pleasant subjective impressions in the viewer and consumer. This optical effect is not possible using the colorants which are currently permitted in the food sector. In contrast to colour
20 pigments which are permitted in the food sector, for example vegetable carbon E153, the titanium dioxide pigments and iron oxide pigments based on platy substrates may be dispersed very readily into the medium to be pigmented. Furthermore, the products thus
25 coloured are distinguished by an increased protection from light and moisture. Vitamin preparations in particular have a longer shelf life. In the case of the colouring of tablets, in many cases a delayed release of active compounds has been observed.

30 It has been found that even at very low amounts of titanium oxide pigments and/or iron oxide pigments novel interesting colours and simultaneously novel properties can be imparted to the foods or
35 pharmaceutical products. Outstanding results are achieved even when the product is coloured with 0.005 to 15.0% by weight, preferably 0.01 to 6.0% by weight, in particular 0.1 to 2.0% by weight of pearlescent pigment, based on the product.

In the event that the pigment is added directly to the food product or pharmaceutical product during manufacture, the amount of pigment used is preferably 0.005 to 4% by weight. In the case of the surface treatment of foods or tablets, the application range is 0.02 to 15.0% by weight, preferably 0.5 to 6.0% by weight, based on the colorant solution or coating solution.

The black iron oxide pigments used are magnetite-coated natural or synthetic mica platelets, talc, kaolin, SiO_2 platelets or TiO_2 platelets. Particularly preferably, finely divided iron oxide pigments are used to colour the products, preferably having a particle size in the range from 0.01 to 200 μm , in particular from 0.1 to 100 μm . Black pearlescent pigments of this type are disclosed, for example, by the patents and patent applications P 23 13 331, P 36 17 430 and JP 90-246314. These pigments are commercially available, for example, under the trademark Candurin® Black Fine from Merck KGaA, Darmstadt.

TiO_2 pigments and Fe_2O_3 pigments based on platy substrates, for example natural and synthetic mica, glass platelets, TiO_2 platelets, SiO_2 platelets and Al_2O_3 platelets are disclosed, for example, by German patents and patent applications DE 14 67 468, DE 19 59 998, DE 20 09 566, DE 22 14 545, DE 22 15 191, DE 22 44 298, DE 23 13 331, DE 25 22 572, DE 31 37 808, DE 31 37 809, DE 31 51 343, DE 31 51 354, DE 31 51 355, DE 32 11 602, DE 32 53 017, WO 93/08237, DE 196 18 564 and EP 0 763 573. The substrates in these patents are coated with one, two, three, four, five or more metal oxide layers. In particular, preference is given to pigments which comprise one or two layers of TiO_2 and/or Fe_2O_3 . The particle sizes of the pigments are preferably $\leq 200 \mu\text{m}$, in particular $\leq 100 \mu\text{m}$.

By adding pearlescent pigments and interference pigments which are permitted for the food sector, and/or natural/nature-identical dyes, colour pigments or colorant fruit extracts and plant extracts, the
5 colouring effect of the titanium dioxide pigments and/or iron oxide pigments can be intensified in the product and simultaneously novel colouring effects can be achieved.

10 In particular, the colouring of food products with a pigment mixture consisting of Fe_3O_4 pigments with pearlescent pigments and/or interference pigments based on mica platelets, Al_2O_3 platelets, SiO_2 platelets or TiO_2 platelets which are coated with TiO_2 and/or Fe_2O_3 ,
15 imparts interesting colouring effects to the products. Very particular preference is given to pigment mixtures comprising an Fe_3O_4 pigment based on mica.

Combining Fe_3O_4 pigments with other pearlescent pigments
20 such as gold pigments, silver pigments or interference pigments intensifies the respective colouring effect of the pigments. This synergy extends considerably the colouring possibilities of the product to be pigmented, without other natural or nature-identical dyes having
25 to be additionally used.

The pearlescent pigments and interference pigments permitted for the food sector are commercially available, for example, under the trademark Candurin®
30 from Merck KGaA.

The total concentration of all pigments in the product to be pigmented should also not exceed 12% by weight, based on the product. The concentration is generally
35 dependent on the specific application. The mixing ratio of the TiO_2 pigments or Fe_3O_4 pigments with a further pigment component depends on the desired effect and is generally 20:1 to 1:20, preferably 5:1, in particular 1:1. The pigment component can be one or more

pearlescent pigments or interference pigments. In particular preference is given to TiO_2 -mica pigments, Fe_2O_3 -mica pigments and TiO_2 - and Fe_2O_3 -coated mica pigments. In addition, preference is given to TiO_2 -
5 and/or Fe_2O_3 -coated or uncoated SiO_2 platelets or TiO_2 platelets.

The colouring effect in food products and pharmaceutical products is improved in particular when
10 black iron oxide pigments are combined with gold pigments, silver pigments and interference pigments based on TiO_2 - or $\text{TiO}_2/\text{Fe}_2\text{O}_3$ -coated mica pigments. The content of Fe_3O_4 pigments in the product should, in this preferred embodiment, preferably be 0.005-2% by weight.

15 In addition to the pearlescent pigments and/or interference pigments, all natural or nature-identical dyes known to those skilled in the art can be added as further colouring component to the titanium dioxide
20 pigments and/or iron oxide pigments. In particular those which may be mentioned here are: E 101, E 104, E 110, E 124, E 131, E 132, E 140, E 141, E 151, E 160a.

25 In addition, other colouring pigments can be added to the platy pearlescent pigments, for example E 171, E 172, E 153.

The content of dyes based on the product is in the
30 range from 0.5 to 25% by weight. Also, fruit extracts and plant extracts can be used as dye, for example carrot juice, beetroot juice, elderberry juice, hibiscus juice, paprika extract, aronia extract.

35 The pharmaceutical products and food products are coloured by adding the titanium dioxide pigment and/or iron oxide pigment to the product to be coloured alone or in combination with other pigments or colorants in the desired quantitative ratios, simultaneously or

successively, during or after their production. Laborious grinding and dispersion of the pigments is not necessary.

- 5 Products suitable for colouring which may be mentioned are in particular coatings on all types of foods, in particular pigmented sugar coatings and shellac coatings (alcoholic and aqueous), coatings containing oils and waxes, containing gum arabic and cellulose
- 10 types (e.g. HPMC = hydroxypropyl methyl cellulose), incorporation into or application onto confectionery, cake decorations, compressed tablets, pan-coated products, chewing gums, gums, fondant products, marzipan products, fillings, cocoa icings and fat
- 15 icings, chocolate and chocolate-containing products, ice cream, cereals, snack products, coating compositions, gateaux presentation plates, hundreds and thousands, sugar crystals, jelly and gelatin products, sweets, liquorice, icing, candyfloss, fat, sugar and
- 20 baker's cream compositions, puddings, desserts, flan glazing, cold sweet soups, beverages containing stabilizing additives such as carboxy methyl cellulose, acidified and non-acidified milk products such as quark, yoghurt, cheese, cheese rinds, sausage casings,
- 25 etc.

- A further large field of use is the pharmaceutical and OTC sector for colouring tablets, gelatin capsules, sugar-coated tablets, ointments, cough syrup, etc. In
- 30 combination with customary coatings such as polymethacrylates and celluloses, for example HPMC, the pigments can be used in many ways for colouring.

- In the case of pan-coated or otherwise coated food
- 35 products and pharmaceutical products, it is possible to combine the titanium dioxide pigments and iron oxide pigments with flavourings (powdered flavourings or liquid flavourings) and/or with sweeteners, for example

aspartame, in order to accentuate the visual effect also in terms of flavour.

5 The invention thus relates to all formulations from the food sector and pharmaceutical sector comprising the titanium dioxide pigment and/or iron oxide pigment alone or in combination with other pigments/pigment mixtures or dyes (natural or nature-identical) as colorants.

10

The examples below are intended to describe the invention without restricting it, however.

Examples

15

Example 1 - Hard caramel manufacturing details

1. Example formula containing Isomalt

20

			Obtainable from:
	Isomalt Merck	75%	Palatinit GmbH,
	Patent GmbH		Mannheim
	Water	24.005%	
	Aspartame	0.0075%	Worlée, Hamburg
25	Acesulfame-K	0.0075%	Nutrinova, Frankfurt a.M.
	Candurin® Silver Sheen ¹	0.144% (0.2% based on the pouring mass)	Merck KGaA, Darmstadt
30	Candurin® Black Fine ²	0.036% (0.05% based on the pouring mass)	Merck KGaA, Darmstadt
	Citric acid	0.5%	Merck KGaA, Darmstadt
35	Flavouring	0.3% (peach 9/030307)	Dragoco, Holzminden

¹ (TiO₂-mica pigment of particle size 5-25 µm)

² (Fe₃O₄-mica pigment of particle size < 15 µm)

Water is heated with Isomalt M to 165°C and the solution is then allowed to cool to 145°C. After addition of citric acid, aspartame, Acesulfame-K, the pigments Candurin® Black Fine and Candurin® Silver Sheen and the flavouring are stirred in. Finally the hot mixture is poured into greased moulds using a pouring funnel. The hard caramels are allowed to cool for 1 hour.

10 Further embodiments:

- It is frequently advisable to dissolve the ingredients in advance using some of the water in order to avoid lump formation.
- The pigments Candurin® Silver Sheen and Candurin® Black Fine can also be heated directly with the water and Isomalt. No loss of colour occurs.

2. Example formula containing Isomalt

20			Obtainable from:
	Isomalt Merck	75%	Palatinit GmbH,
	Patent GmbH		Mannheim
	Water	24.041%	
	Aspartame	0.0075%	Worlée, Hamburg
25	Acesulfame-K	0.0075%	Nutrinova,
			Frankfurt a.M.
	Candurin®	0.144% (0.2% based	Merck KGaA,
	Honeygold ³	on the	Darmstadt
		pouring mass)	
30	Citric acid	0.5%	Merck KGaA,
			Darmstadt
	Flavouring	0.3% (peach 9/030307)	Dragoco,
			Holzminde
	³ (TiO ₂ /Fe ₂ O ₃ -mica pigment of particle size 5-25 µm)		

35

Water is heated with Isomalt M to 165°C and the solution is then allowed to cool to 145°C. After addition of citric acid, aspartame, Acesulfame-K, Candurin® Honeygold and the flavouring are stirred in.

Finally, the hot mass is poured into greased moulds using a pouring funnel. The hard caramels are allowed to cool for 1 hour.

5 Further embodiments:

- Frequently it is advisable to dissolve the ingredients in advance using some of the water in order to prevent lump formation.
- The pigment Candurin® Honeygold can also be heated directly with the water and Isomalt.

3. Example formula containing sugar

			Obtainable from:
15	Sugar	41%	Südzucker
	Water	17.077%	
	Glucose syrup	41% C [*] Sweet	Cerestar, Krefeld
	Candurin® Silver	0.082% (0.1% based	Merck KGaA,
	Sheen ¹	on the	Darmstadt
20		pouring mass)	
	Candurin® Black	0.041% (0.05% based	Merck KGaA,
	Fine ²	on the	Darmstadt
		pouring mass)	
	E 104 1:100 dilution	0.4% Sikovit	BASF,
25	Flavouring	0.4% (banana 9/030388)	Ludwigshafen
			Dragoco,
			Holzminde

¹ (TiO₂-mica pigment of particle size 5-25 µm)

² (Fe₃O₄-mica pigment of particle size < 15 µm)

30

The sugar is heated with the water to 100°C and then glucose syrup is added. The solution is then heated to 145°C. After addition of Candurin® Silver Sheen, Candurin® Black Fine, dye solution and the flavouring, the caramel solution is poured into greased moulds using a pouring funnel. Finally, the mixture is allowed to cool for two hours. The Candurin® pigments can be

35

the glucose syrup. This variant contains no acid, since this would make the caramelization too intense.

4. Example formula containing sugar

5

			Obtainable from:
	Sugar	41%	Südzucker
	Water	17.118%	
	Glucose syrup	41% C [*] Sweet	Cerestar, Krefeld
10	Candurin [®] Wine Red ⁴	0.082% (0.1% based on the pouring mass)	Merck KGaA, Darmstadt
	E 104 1:100 dilution	0.4% Sikovit	BASF, Ludwigshafen
15	Flavouring	0.4% (banana 9/030388)	Dragoco, Holzminden
	⁴ (Fe ₂ O ₃ -mica pigment of particle size 10-60 µm)		

- 20 The sugar is heated with the water to 100°C and then glucose syrup is added. The solution is then heated to 145°C. After addition of Candurin[®] Wine Red, dye solution and the flavouring, the caramel solution is poured into greased moulds using a pouring funnel. Finally, the mixture is allowed to cool for two hours.
- 25 The Candurin[®] pigment can be either mixed with the sugar or added as a mixture with the glucose syrup. This variant contains no acid, since this would make the caramelization too intense.

30 Example 2 - Manufacture of gelatin articles

1. Example formula

			Obtainable from:
35	Water	10.48%	
	Sugar	31.45%	Südzucker
	Glucose syrup	31.45% C [*] Sweet	Cerestar, Krefeld
	Candurin [®] Wine Red	0.38% (0.4% based on the	Merck KGaA, Darmstadt

pouring mass)

	Citric acid 1:1	2.51%	Merck KGaA,
	dilution		Darmstadt
	Gelatin	7.86% 260 Bloom	DGF, Eberbach
5	Water	15.748%	
	Flavouring	0.122% (blackcurrant	Dragoco,
		9/695750)	Holzminen

10 The gelatin is first softened with twice the amount of water at 60°C. Sugar and water are heated to 100°C, then the glucose syrup is added. The mixture is heated further to 120°C and is then allowed to cool to approximately 85°C. The Candurin® pigment, the citric acid, the flavouring and the gelatin solution are

15 stirred in, and the deaerated gelatin mixture is charged into greased moulds using the pouring funnel. The product is allowed to cool for approximately 16 hours.

20 Further embodiments:

- The Candurin® pigment can here again be already mixed directly with the sugar or introduced together with the glucose syrup.
 - Instead of pouring into moulds, the traditional
- 25 method using negative moulds in moulding powder can be used in this case to produce gelatin articles.

2. Example formula

30			Obtainable from:
	Water	10.508%	
	Sugar	31.45%	Südzucker
	Glucose syrup	31.45% C' Sweet	Cerestar, Krefeld
	Candurin® Blueberry	0.38%	(0.4% based Merck KGaA,
35	Sugar ⁵	on the	Darmstadt
		pouring mass)	
	E 153 (vegetable	0.038%	(0.04% based Dr. Marcus
	carbon/Carbon	on the	
	medicinales)	pouring mass)	

	Citric acid 1:1	2.51%	Merck KGaA,
	dilution		Darmstadt
	Gelatin	7.86% 260 Bloom	DGF, Eberbach
	Water	15.682%	
5	Flavouring	0.122% (blackcurrant	Dragoco,
		9/695750)	Holzminde

*5 (TiO₂-mica pigment of particle size 10-60 µm)

The gelatin is first softened with the same amount of water at 60°C. Sugar and water are heated to 100°C, then the glucose syrup is added. The mixture is heated further to 120°C and is then allowed to cool to approximately 85°C. The Candurin® pigment, the citric acid, the flavouring and the gelatin solution are stirred in, and the deaerated gelatin mixture is charged into greased moulds using the pouring funnel. The product is allowed to cool for approximately 16 hours.

20 Further embodiments:

- The Candurin® pigment can here again be already mixed directly with the sugar or introduced together with the glucose syrup.
- Instead of pouring into moulds, the traditional method using negative moulds in moulding powder can be used in this case to produce gelatin articles.

Example 3 - Dragees

30 a) Cores to be coloured: white dragee cores (liquorice rods with a hard sugar coating)

Solution for coating the dragees:

- | | | |
|----|---|-----------------------------|
| | | Obtainable from: |
| 35 | - alcoholic shellac solution | 95.62% Wolff & Olsen |
| | - Candurin® Silver Lustre* ¹ | 4.00% Merck KGaA, Darmstadt |
| | - Candurin® Black Fine* ² | 0.38% Merck KGaA, Darmstadt |

*1 (TiO₂-mica pigment of particle size 10-60 µm)

*2 (Fe₃O₄-mica pigment of particle size < 15 µm)

The Candurin® pigments are evenly distributed in the shellac and sprayed onto the dragees which are slowly rotating in the dragee pan. The dragees are
5 continuously dried using cold air. The spraying is continued until the desired colour coverage is achieved. Finally, the cores are taken out of the pan and dried on racks for approximately 12 hours.

- 10 b) Cores to be coloured: black dragee cores (chewing gum balls)

Solution for coating the dragees:

- Obtainable from:
- 15 - alcoholic shellac solution 95% Kaul GmbH
- Candurin® Caramel* 5% Merck KGaA, Darmstadt
* (Fe₂O₃-mica pigments of particle size 10-60 µm)

The Candurin® pigment is evenly distributed in the shellac and sprayed onto the dragees which are slowly rotating in the dragee pan. Spraying is continued until the desired colour coverage is achieved. Drying is performed continuously using cold air in order to prevent the cores from sticking together. Finally, the
20 cores are taken out of the pan and dried on racks for approximately 12 hours.

- c) - Coating liquorice nibs with a white hard sugar shell

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Cores to be coloured: liquorice nibs having a white hard sugar shell

Solution for coating the dragees:

- Obtainable from:
- 35 - alcoholic shellac solution 95.75%
Capol 425 Kaul GmbH
- Candurin® Buttergold*¹ 4% Merck KGaA, Darmstadt
- Candurin® Black Fine 0.25% Merck KGaA, Darmstadt

*¹ (TiO₂/Fe₂O₃-mica pigment of particle size 10-60 μm)

The cores are coated in a similar manner to Example 3 a).

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d) - Pan-coating of Viennese nuts (white, hard sugar shell with hazelnut core)

Solution for coating the dragees:

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Obtainable from:

- alcoholic shellac solution 96.0% Wolff & Olsen
- Candurin® Silver Lustre* 4% Merck KGaA, Darmstadt
- * (TiO₂-mica pigment of particle size 10-60 μm)

15

The Candurin® pigment is distributed evenly in the shellac solution. It is applied in a similar manner to Example 3 a).

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e) - Starting material: white chocolate drops

An aqueous Candurin®/HPMC solution is used for the spray application.

25

- The Candurin® pigments are stirred into water. Then, if appropriate, additional dyes, flavourings or sweeteners are then added. Finally, the film-forming agent (HPMC) is added to the suspension. As a result of the increasing viscosity, the stirrer speed must also be correspondingly increased. After approximately 40-60 minutes, the HPMC is completely dissolved and the solution can then be sprayed onto the dragees.

30

- The spray application can be performed in the pan-coating drum or in customary coating systems, with care being taken to ensure an appropriately matched continuous drying air stream (temperatures/volumes).

35

- As soon as the desired colouring effect is achieved, the dragees are discharged. They are then dried on racks.

Spray solution:

Sepifilm Lp10	6.0%	Seppic
Candurin® paprika	5.0%	Merck KGaA, Darmstadt
Flavouring	0.5% (vanilla 9/024233)	Dragoco, Holzminden
Water	88.5%	

(Fe₂O₃-mica pigment of particle size 10-60 µm)

- 5 f) Starting material: white sugar-pan-coated almonds

The spray solution is prepared and applied as in e)

Spray solution:

Sepifilm Lp10	5.0%	Seppic
Candurin® Silver Lustre	4.0%	Merck KGaA, Darmstadt
Sepisperse M5062	1.0%	Seppic
Water	90.0%	

- 10 (TiO₂-mica pigment of particle size 10-60 µm)

- g) Starting material: white sugar-pan-coated almonds

The spray solution is prepared and applied as in e)

15

Spray solution:

Sepifilm Lp10	6.0%	Seppic
Candurin® Silver Lustre	4.0%	Merck KGaA, Darmstadt
El53	0.2%	Dr. Marcus
Water	89.8%	

(TiO₂-mica pigment of particle size 10-60 µm)

- h) Starting material: red sugar-pan-coated almonds

20

An aqueous Candurin®/gum arabic solution is used for the spray application.

The Candurin® pigments are stirred into the gum arabic solution and are then sprayed onto the dragees rotating

25

in the pan-coating drum. Drying should be performed continuously here using cold air. When the desired colouring effect is achieved, the spraying operation is terminated. The coloured dragees can then further be coated with a shellac film to avoid sticking together.

Spray solution:

Gum arabic solution	82.0%	Kaul
Candurin® Wine Red*	8.0%	Merck KGaA, Darmstadt
Water	10.0%	

* (Fe₂O₃-mica pigment of particle size 10-60 µm)

10 Example 4 - Jelly dessert

		Obtainable from:
	Calcium lactate	0.05% Merck KGaA
	Genugel LC4N	0.6% Hercules
15	Tetrasodium phosphate	0.15% Merck KGaA
	Citric acid crystalline	0.38% Merck KGaA
	gellan	0.06% Kelco
	Tripotassium phosphate	0.05% Merck KGaA
	Sucrose	15% Südzucker
20	Candurin® Silver Sparkle ¹	0.025% Merck KGaA
	Flavouring and colour optional	
	Water	83.685%

¹ (TiO₂-mica pigment of particle size 20-150 µm)

25 All components are heated to 95°C and kept at this temperature for 3-5 minutes with stirring. They are then allowed to cool with gentle stirring to 40-45°C. The jelly dessert is then poured into moulds and if necessary it is cooled.

30

Pharmaceutical applications:

Example 5

- 5 a) Initial weight 1 kg of white tablets $d = 8 \text{ mm}$,
 $G = 200 \text{ mg}$

Solution for film coating:

6%	Sepifilm Lp10 (Mixture of hydroxypropyl methyl cellulose, stearic acid and microcrystalline cellulose)	Seppic
5%	Candurin® Caramel'	Merck KGaA, Darmstadt
89%	Water	

* (Fe_2O_3 -mica pigment of particle size 10-60 μm)

10

Total amount applied: 200 g
 This corresponds to 1.2 mg of polymer/ cm^2 of tablet
 surface area

- 15 b) Initial weight 1 kg of white tablets $d = 8 \text{ mm}$,
 $G = 200 \text{ mg}$

Solution for film coating (100 g):

6%	Sepifilm Lp10	Seppic
4%	Candurin® Silver Lustre'	Merck KGaA
1%	Sepisperse M5062 (Dye paste: TiO_2 , natural or nature- identical dyes, stabilizer)	Seppic
89%	Water	

* (TiO_2 -mica pigment of particle size 10-60 μm)

20

Total amount applied: 200 g
 This corresponds to 1.2 mg of polymer/ cm^2 of tablet
 surface area

- 25 Production of the film-coating solution:

- The Candurin® pigments are stirred into water. If appropriate, additional dyes are then added. Finally, the film-forming agent (HPMC) is dispersed in the suspension. The increasing viscosity necessitates the stirrer viscosity also being correspondingly increased. After approximately 40-60 minutes, the HPMC is completely dissolved and the solution can then be sprayed onto the tablets.
- The spray application is made using a customary standard coating process.

Sugar confectionery

Example 6:

a) Liquorice products:

Raw material: extruded liquorice products

- The oiled liquorice products are sprayed with a Candurin®/shellac solution in a pan-coating drum. At the same time they are dried using cold air. As soon as the desired colouring effect is achieved, application is halted and the coloured liquorice products are discharged from the pan.

Spray solution:

Alcoholic shellac solution	97%	Kaul
Candurin® Kiwi Sugar*	3%	Merck KGaA, Darmstadt

* (TiO₂-mica pigment of particle size 10-60 µm)

b) Menthol pastilles

Raw material: menthol pastilles

- A Candurin®/shellac solution (aqueous) is sprayed onto the rotating pastilles in the pan-coating drum. Drying is performed continuously with warm air here. As soon as the desired colouring effect is achieved,

application is terminated and the coloured pastilles are discharged from the pan.

Spray solution:

Alcoholic shellac solution	96%	Warner Jenkinson
Candurin® Silver Lustre*	4%	Merck KGaA, Darmstadt

5 * (TiO₂-mica pigment of particle size 10-60 µm)

c) Marzipan roses (cake decoration)

Raw material: red marzipan roses

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The cake decorations are sprayed with a Candurin®/shellac solution until the desired colour application is achieved. Subsequent drying with cold air is possible.

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Spray solution:

Alcoholic shellac solution	97%	Wolff & Olsen
Candurin® Blueberry Sugar*	3%	Merck KGaA, Darmstadt

* (TiO₂-mica pigment of particle size 10-60 µm)

d) Sherbet sweets

20

Raw material: sherbet sweets, white

The sherbet sweets are sprayed with a Candurin®/shellac solution until the desired colour application is achieved. Subsequent drying with cold air is possible.

25

Spray solution:

Alcoholic shellac solution	94%	Kaul
Candurin® Silver Sparkle*	6%	Merck KGaA, Darmstadt

* (TiO₂-mica pigment of particle size 20-150 µm)

Patent claims

1. Use of titanium dioxide pigments and/or iron oxide pigments based on platy substrates for colouring food products or pharmaceutical products.
2. Use of titanium dioxide pigments and/or iron oxide pigments according to Claim 1, characterized in that the platy substrate is a mica platelet, Al_2O_3 platelet, TiO_2 platelet or SiO_2 platelet.
3. Use of titanium dioxide pigments and/or iron oxide pigments according to Claim 1 or 2, characterized in that the iron oxide pigment is a platy substrate coated with Fe_2O_3 or Fe_3O_4 .
4. Use of titanium dioxide pigments and/or iron oxide pigments according to one of Claims 1 to 3, characterized in that the titanium dioxide pigment content and/or iron oxide pigment content in the food product or pharmaceutical product is 0.005 to 15% by weight.
5. Use of titanium dioxide pigments and/or iron oxide pigments according to one of Claims 1 to 4, characterized in that the titanium dioxide pigment and the iron oxide pigment can be mixed in a ratio of 1:20 to 20:1.
6. Use of titanium dioxide pigments and/or iron oxide pigments according to one of Claims 1 to 5, characterized in that titanium dioxide pigments and/or iron oxide pigments combined with one or more pearlescent pigments, coated or uncoated TiO_2 platelets, SiO_2 platelets, natural or nature-identical colorants, colour pigments or natural plant or fruit extracts are used.

7. Use of titanium dioxide pigments and/or iron oxide pigments according to Claim 6, characterized in that the titanium dioxide pigments and/or iron oxide pigments are used in combination with one or more pearlescent pigments based on mica coated with TiO_2 , Fe_2O_3 or a $\text{TiO}_2/\text{Fe}_2\text{O}_3$ mixture.
8. Use of titanium dioxide pigments and/or iron oxide pigments according to Claim 6 or 7, characterized in that the iron oxide pigment is a mica platelet, SiO_2 platelet or TiO_2 platelet coated with Fe_3O_4 .
9. Use of titanium dioxide pigments and/or iron oxide pigments according to one of Claims 1 to 8, characterized in that the food product or pharmaceutical product is furnished with a coating of shellac, oils, waxes, gum arabic, celluloses, polymethacrylates or icing comprising titanium dioxide pigments and/or iron oxide pigments and if appropriate other pigments and/or colorants.
10. Process for the production of food products and pharmaceutical products coloured by titanium dioxide pigments and/or iron oxide pigments, characterized in that the titanium dioxide pigment and/or iron oxide pigment is added to the product to be coloured alone or in combination with other pigments or colorants in the desired quantitative ratios, simultaneously or successively, during or after their production.
11. Food products and pharmaceutical products comprising titanium dioxide pigments and/or iron oxide pigments based on platy substrates as colorant.

Abstract

The present invention relates to the use of titanium dioxide pigments and/or iron oxide pigments based on platy substrates for colouring food products and pharmaceutical products.

Docket No.
Merck

Declaration and Power of Attorney For Patent Application

English Language Declaration

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name,

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

the specification of which

(check one)



is attached hereto.



was filed on 08.07.1999 as United States Application No. or PCT International Application Number PCT/EP99/04792 and was amended on _____

(if applicable)

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose to the United States Patent and Trademark Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, Section 1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, Section 119(a)-(d) or Section 365(b) of any foreign application(s) for patent or inventor's certificate, or Section 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate or PCT International application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application(s)

Priority Not Claimed

198 31 869.3
(Number)

Germany
(Country)

16.07.1998
(Day/Month/Year Filed)



(Number)

(Country)

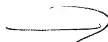
(Day/Month/Year Filed)



(Number)

(Country)

(Day/Month/Year Filed)



I hereby claim the benefit under 35 U.S.C. Section 119(e) of any United States provisional application(s) listed below:

(Application Serial No.)

(Filing Date)

(Application Serial No.)

(Filing Date)

(Application Serial No.)

(Filing Date)

I hereby claim the benefit under 35 U.S.C. Section 120 of any United States application(s), or Section 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States of PCT International application in the manner provided by the first paragraph of 35 U.S.C. Section 112. I acknowledge the duty to disclose to the United States Patent and Trademark Office all information known to me to be material to patentability as defined in Title 37, C.F.R., Section 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application:

(Application Serial No.)

(Filing Date)

(Status)
(patented, pending, abandoned)

(Application Serial No.)

(Filing Date)

(Status)
(patented, pending, abandoned)

(Application Serial No.)

(Filing Date)

(Status)
(patented, pending, abandoned)

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

→

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